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COMPLETE SPECIFICATION

Method for Producing Fine Crystals of Controlled Particle Size

I, THE MINISTER OF NATIONAL DEFENCE OF HER MAJESTY'S CANADIAN GOVERNMENT, of the City of Ottawa, in the Province of Ontario, Canada, do hereby declare the invention, for which I pray that a patent may be granted to me, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to a method for the precipitation from solution of crystalline materials to obtain a fine crystalline product of controlled particle size. The invention relates particularly to the production of fine crystalline cyclotrimethylenetrinitramine commonly referred to as RDX, in a particle size which is both uniform and readily reproducible.

RDX has recently been proposed as an ingredient in propellant explosives. However since RDX is a crystalline material, its satisfactory incorporation in propellants presents a number of problems not met with in the case of more conventional materials such as nitro-glycerine and nitro-cellulose. Uniformity of particle size is a practical necessity in obtaining uniformity in ballistic performance and further, in order to provide the highest practical density, the incorporated crystalline material should have as fine a particle size as may be feasibly reproduced with uniformity. Proposals have been made for producing RDX of controlled particle size by precipitation from an acetone solution by the addition of water. Accordingly, fine RDX has been prepared by the rapid drowning of hot RDX-acetone solution in excess water with rapid stirring. When manufactured in this manner, the product will normally vary in particle size over a wide range, say from 2—20μ. It is then necessary to sieve the material to remove the oversize and produce a product which meets the specification which is being adhered to. It has been found impossible to eliminate variation in particle size using conventional precipitation methods and conventional apparatus. This is due primarily to the fact that conditions of agitation and mixing of the acetone solution and the precipitating water cannot be made constant and cannot be perfectly reproduced from one operation to the next.

I have now found that the above difficulties can be eliminated and that fine RDX of controlled particle size can be produced with excellent reproducibility if the RDX-acetone solution and the precipitating water are each projected as jets and caused to impinge upon each other at an angle of from about 60° to about 90°. The mixing thus produced is substantially instantaneous, and provided that the crystalline product is removed from the mother liquor without undue delay, no opportunity is afforded for crystal growth following initial precipitation.

According to the invention, therefore I provide a process for the production of fine, uniform crystals of RDX, comprising forming a first jet of liquid consisting of a solution of RDX in acetone, forming a second jet of water which is maintained at a temperature of less than about 30° C. the mass ratio of said first jet to said second jet being maintained at a value less than about 3:1, directing said jets so that they impinge upon each other at an angle from about 60° to 90°, whereby the RDX is precipitated from said solution; and separating the thus precipitated RDX from the thus resulting mother liquor. The final separation is preferably done in a centrifuge.

The particle size of the precipitate may be controlled by varying the concentration of the RDX in acetone solution, and also by maintaining the temperature of the precipitant water at a value below about 30° C.

In a further embodiment of the invention, a desensitizing or phlegmatizing agent may be added to the RDX in acetone solution and precipitated from solution as a protective film on the crystals of RDX; thus rendering the material less hazardous to handle and transport.

The apparatus for carrying out the process according to the invention comprises at least one pair of jet nozzles mounted in converging relationship so that streams issuing from them will impinge at an angle from about 60° to about 90°, means for supplying RDX-acetone solution to one nozzle of said pair at a controlled rate of flow and concentration, means for supplying precipitating water to the other nozzle of said pair at a controlled rate of flow,

means for controlling the temperature of the liquids passing through said apparatus and means for collecting the resulting mother liquor and precipitate and recovering the precipitate.

The nozzles used may have any desired size of orifice up to about 0.5 inches, so that a wide range of values of the output capacity can be obtained.

The invention and its operation will be more fully understood from a reading of the following detailed description in conjunction with the accompanying drawings wherein:—

Figure 1 is a perspective view of the mixer head in an apparatus for carrying out the invention showing a typical arrangement of pairs of jet nozzles,

Figure 2 is a vertical section of the apparatus illustrated in Figure 1 taken along the plane 2—2, and

Figure 3 is a diagrammatic flow sheet of the process according to the invention.

The mixer head illustrated comprises coaxial chambers 10 and 11, adapted near their upper portions for joining to inlet ducts 12 and 13 respectively, by suitable means such as threaded unions, and having near their bottom portion a plurality of outlet ports, the outlet ports of chamber 10 being adjacent to and in one to one correspondence with the outlet ports of chamber 11. The outlet ports are adapted to receive threaded nozzles 14 and 15 in such a manner that the longitudinal axes of each pair of nozzles intersect exterior to the mixer head at an angle α which may be from about 60° to about 90°.

The coaxial chambers 10 and 11 are enclosed by an outer jacket 16 adapted for connection by suitable means, such as threaded unions, to an inlet duct 17 and an outlet duct 18.

The flow sheet for the process shows the mixer head 20 connected through an insulated duct 21, which is fitted with a flow meter 22 and a flow regulating valve 23, to a source of supply 24 of the pregnant solution; and connected through a second insulated duct 25, flow meter 26, and flow regulating valve 27, to a source of supply 28 of the precipitant fluid.

The components of this apparatus may be constructed of any suitable material, provided it is chemically inert to the materials used in the process.

In the operation of this invention a hot filtered solution of RDX in acetone is fed to the mixer head through a calibrated flow meter 22 via the duct 21, to the inlet 13. Precipitant water is fed to the mixer head through a similar calibrated flow meter 26 via the duct 25 to the inlet 12. A flow of steam or hot water is maintained in the outer jacket 16 to prevent possible crystallization of the RDX solution in the chamber 11 and subsequent blocking of the outlets. The streams or jets of RDX solution and water impinge upon each other and are thus rapidly and intimately mixed. This rapid mixing causes a myriad of minute crystallization nuclei to be formed, and, since the intimate nature of the mixing obviates varying concentrations of RDX solution in the mixture each of the nuclei tends to grow to a uniform size until the solution is depleted of the precipitating material. This factor affords control of particle size, in that larger crystals will result when the mixture contains more of the precipitating material.

By varying the volumetric flow in one or both of the effluent jets, effective control over a wide range of the mass ratio of the RDX-acetone solution to water is obtained. The results of extensive experimentation on the effect of varying the mass ratio are tabulated below. The RDX-acetone solution is a 10% solution except where indicated. The nozzle holes are of 1/16″ diameter.

TABLE I.

Mass ratio of RDX-Acetone/water	Temp. of water °C	Total feed gms./min.	Crystal size μ
0.107:1	13	3100	1 and less
0.15 :1	15	2300	1—2
0.125:1	4	2700	1—2
0.125:1	10	2700	1—2
0.125:1	15	2700	1—2
0.125:1	32	3240	4—5 tending to rods
0.125:1	50	2700	3—20 rods
*0.125:1	15	2700	1—2
0.20 :1	4	1820	1—2
0.21 :1	13	3380	1—2
0.23 :1	15	2825	1—2
0.31 :1	10	1200	1—2

*RDX-Acetone-five percent solution.

TABLE II.

Mass ratio of RDX-Acetone/water	Temp. of water °C	Total feed gms./min.	Crystal size μ
0.425:1	14	2850	2—8
0.5 :1	13	3200	2—3
0.7 :1	14	3400	2—10
0.92 :1	15	3000	5—10
1 :1	15	4000	3—10
1.5 :1	15	4000	8—25
2 :1	13	3600	5—20
3 :1	15	3450	8—30

Table I indicates the range of optimum conditions for the production of very fine uniform crystals. It will be noted that the temperature of the water used as precipitant is not critical, except at temperatures somewhat above normal room temperature. Under the latter conditions, the temperature coefficient of solubility slows the rate of precipitation allowing preferential growth of the larger crystals, thus giving a wider range of particle size.

Table II indicates that for mass ratios of RDX-acetone to water between 0.3:1, and 3:1 a product of larger crystals over wider ranges of particle size is obtained under similar operating conditions as the mass ratio is increased.

Once the precipitation has been effected, the precipitate and mother liquor are collected by a funnel and run to a separating device. To obviate the possibility of further crystal growth in the mother liquor this separation must necessarily be made as quickly as possible. For the purpose of experimentation, filtration by means of a centrifuge was found convenient. However, for the purpose of a continuous process any other method of filtering by the application of pressure of suction may be used. One such method, for example, would be to collect the crystalls on a filter screen travelling over a suction box and subsequently to remove the crystals from the screen by a doctor.

The effect of varying the nozzle size is shown in Table III.

TABLE III.

Mass ratio of RDX-Acetone/water	Nozzle holes inches	Temp. of water °C.	Total feed gms./min.	Crystal size μ
0.125:1	1/16	10	2700	1—2
	3/32	9	2700	1—2
	5/32	10	2700	1—2
0.5 :1	1/16	13	3200	2—3
	3/32	9	2400	2—5
	5/32	10	2400	2—5
1 :1	1/16	15	4000	3—10
	3/32	9	4000	2—10
	5/32	10	4000	5—10
2 :1	1/16	13	3600	5—20
	3/32	9	3600	10—50
	5/32	10	3600	15—60
3 :1	1/16	15	3450	8—30
	3/32	9	3240	20—65
	5/32	10	3240	15—100

It is seen that for low mass ratio of RDX-acetone to water the effect of increasing the nozzle size is not particularly significant. Very fine crystals (1—2μ) are formed using nozzles with holes from 1/16″ diameter to 5/32″ diameter under similar conditions of mass ratio, feed rate and temperature. However, at mass ratios greater than 1:1, larger crystals and wider ranges of particle size are obtained as the size of the nozzle hole is increased. Experiments have also been carried out using nozzles with 1/2″ diameter holes. Although the finest size crystals were not obtained using these larger jets, it was possible to prepare fine crystals (2—10μ) with regularity and reproducibility. As before, the crystal sizes can be controlled by adjusting the mass ratio of the feed.

With the smaller units, the mixer head is capable of producing about 10 pounds per hour of the fine (1—2μ) RDX; with a pair of

the 1/2" nozzles an output of 4 pounds RDX (2—10μ) per minute is possible. Clearly a battery of mixer heads could be operated efficiently to obtain any desired production capacity.

A further desirable feature of this invention relates to desensitizing RDX to impact detonation. It is well known that the addition of small amounts (about 10%) of wax to crystalline explosive compounds will render them less sensitive to impact. The general method of achieving this is to add the molten wax to an agitated slurry of the material in excess hot water.

In the present invention small quantities of beeswax or petroleum wax can be added to the hot RDX-actone solution. When the RDX is precipitated as crystals from such a solution by the process hereinbefore described, each crystal is coated with a thin protective film of the wax. The amount of wax thus precipitated may be from 5 to 8%. With the very fine crystals (up to 10μ) the sensitiveness to impact has been decreased by a factor of 2 and for larger crystals (10μ—75μ) the sensitiveness is decreased by a factor of 3. Thus, a phlegmatized form of the explosive RDX can be obtained without additional processing and, therefore, without need of handling the material in a sensitive form.

What we claim is:—

A process for the production of fine, uniform crytals of RDX, comprising forming a first jet of liquid consisting of a solution of RDX in acetone, forming a second jet of water which is maintained at a temperature of less than about 30° C., the mass ratio of said first jet to said second jet being maintained at a value less than about 3:1, directing said jets so that they impinge upon each other at an angle from about 60° to 90°, whereby the RDX is precipitated from said solution; and separating the thus precipitated RDX from the thus resulting mother liquor.

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741,756 COMPLETE SPECIFICATION
1 SHEET
This drawing is a reproduction of the Original on a reduced scale.

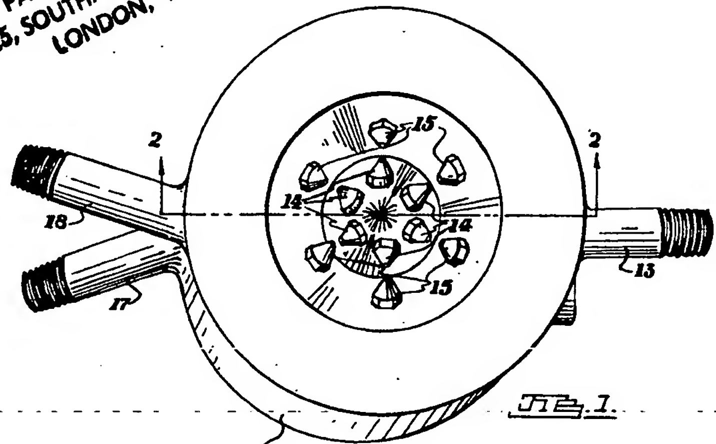

Fig. 1.

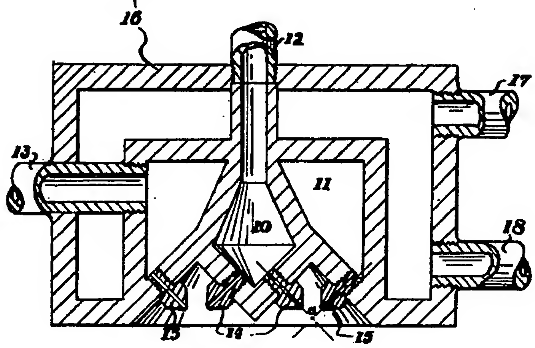

Fig. 2.

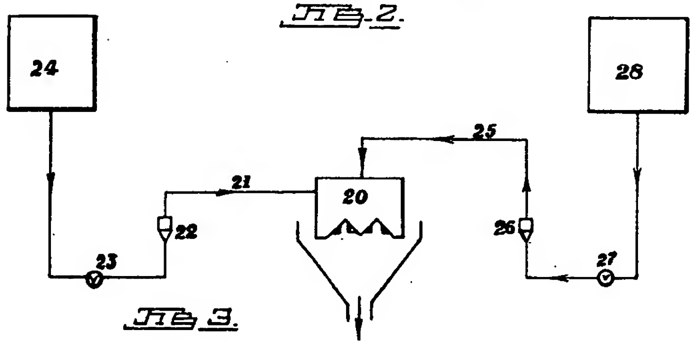

Fig. 3.